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Static Magnetic Ordering of CeCu_{2.1}Si₂ Found by Muon Spin Relaxation

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Zero- and longitudinal-field muon spin relaxation measurements on a poly-crystal sample of a heavy-fermion superconductor CeCu_{2.1}Si₂ ($T_C = 0.7K$) have revealed an onset of static magnetic ordering below $T \sim 0.8K$. The line shapes of the observed spectra in zero field indicate a wide distribution of static random local fields at muon sites, suggesting that the ordering is either spin glass or incommensurate spin-density-wave state. The observed width of the random local field at $T = 0.05K$ corresponds to a small averaged static moment of the order of $0.1\mu_B$ per formula unit.

CeCu₂Si₂ is the first heavy-fermion system which was found to become superconducting at $T_C \sim 0.5K$ (ref. 1). Although there were signatures suggesting possible magnetic orderings in non-superconducting CeCu_{1.9}Si₂ (ref. 2) and Ce_{1-y}La_yCu₂Si₂ with $y \geq 0.2$ (ref. 3), superconducting specimens CeCu_xSi₂ with $x = 2.0 \sim 2.2$ have so far been believed to have purely superconducting ground states without magnetic ordering. In this paper, we present direct evidence from muon spin relaxation (μ SR) measurements that superconducting CeCu_{2.1}Si₂ ($T_C = 0.7K$) undergoes a random static magnetic ordering below $T \sim 0.8K$.

It is known that a small amount of off-stoichiometric excess Cu helps to stabilize the superconductivity of CeCu₂Si₂. Therefore, we prepared a polycrystal specimen of CeCu_{2.1}Si₂ with the method described in ref. 4. The superconducting transition temperature $T_C = 0.7K$ was determined by a resistivity measurement on a small piece cut out from the present specimen. A neutron scattering measurement on the crystal structure confirmed that there is no minor phase within the accuracy of a few volume percent.

Zero- and longitudinal-field μ SR measurements were performed using polarized positive muon beams at AGS (BNL) and SIN (Zürich) muon channels. In the zero-field measurements, very small depolarization of muon spins was observed above $T \approx 0.9K$, while the depolarization rate increased rapidly with decreasing temperature below $0.8K$. The zero-field muon spin relaxation functions $G_z(t)$ (ref. 5) observed at different temperatures show no precession signal but have line shapes similar to those observed in dilute-alloy spin glasses⁵. This indicates that the magnitude of local fields at muon sites varies widely, in contrast to the cases for uniform ferro- or antiferromagnets.

A phenomenological form for the muon spin relaxation function

$$G_z(t) = \frac{A_1}{A} \exp(-\frac{1}{2}\sigma^2 t^2) + \frac{A_2}{A} \exp(-\Lambda t), \quad (1)$$

with $A_1/A \sim 2/3$ and $A_2/A \sim 1/3$, gives good fits to all the data observed in zero-field. The first (second) term of eq. (1) corresponds to the depolarization of muon spins by the components

of internal fields perpendicular (parallel) to the initial polarization direction of the muon spins. Figure 1 shows the temperature dependence of the relaxation rate σ .

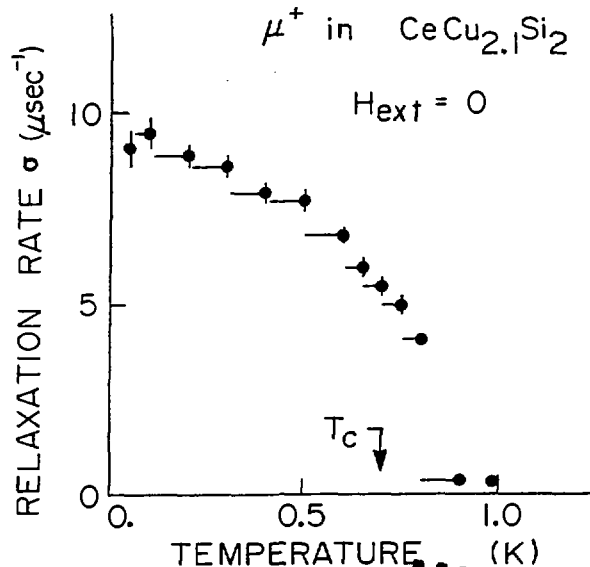


Fig. 1. Muon spin depolarization rate σ , as defined in eq. (1), derived from the relaxation functions observed in CeCu_{2.1}Si₂ in zero field. The onset of magnetic ordering is seen around $T_M \sim 0.8K$. The superconducting transition temperature T_C , determined by a resistivity measurement on a piece cut off from the present specimen, is indicated by the arrow.

In order to distinguish whether this depolarization is due to static or fluctuating local fields, we have also performed μ SR measurements by applying longitudinal external magnetic fields $H_L = 250G$ and $1kG$ parallel to the initial muon spin direction. The muon spin polarization had a finite value $G_s(t) \sim 0.45$ with $H_L = 250G$, and $G_s(t) \sim 0.95$ with $H_L = 1kG$, almost independent on time between $0.5\mu sec \leq t \leq 2\mu sec$ at $T = 0.1K$. A similar decoupling of the random field was observed around $T = 0.8K$. These results indicate that the depolarization shown in Fig. 1 is due predominantly to the static random local fields. The rapid increase of σ below $T \sim 0.8K$ then corresponds to the sharp onset of static magnetic ordering around the ordering temperature $T_M = 0.8K$. Due to the limited accuracy of the temperature measurements with the cold-finger dilution cryostat used in the present experiment, it is not clear whether the magnetic and superconducting orderings occur simultaneously at the same temperature or independently at different temperatures. It is, however, evident that the superconductivity and magnetic ordering coexist below $T_C = 0.7K$.

The wide distribution of the static random local fields, as observed in the present experiment, can be expected either for spin glass (SG) or incommensurate spin-density-wave (ISDW) systems. The spin glass ordering is consistent with a large field dependence of the susceptibility observed in the non-superconducting system $CeCu_{1.9}Si_2$ (ref.2). If one assumes that a majority of Ce (or Cu) moments participate in the spin-glass freezing, the zero-field relaxation rate $\sigma \sim 10\mu sec^{-1}$ (the width $\sigma/\gamma_\mu \sim 120G$ of the local field) observed at $T = 0.05K$ corresponds to the dipolar field from a static moment of the order of 0.1 Bohr magneton per formula unit. If the small population of the Ce^{3+} ions with an ordered moment of $5\mu_B$ forms a spin glass, this value of σ is expected for the freezing moments on a few percent of the entire Ce atoms. From the present work alone, it is not possible to distinguish among the above-mentioned three possible spin structures (the two types of SG states and the ISDW state) of $CeCu_{2.1}Si_2$ below T_M .

Recently, a heavy-fermion superconductor UPt_3 ($T_C \sim 0.5K$) was found to order magnetically below $T \sim 5K$ with an extremely small averaged moment of $0.001 \sim 0.02\mu_B/U$ (refs 6,7). URu_2Si_2 is another superconducting heavy-fermion system ($T_C \sim 1.0K$) which orders antiferromagnetically below $T_N \sim 17K$ with an ordered moment of $0.03\mu_B$ per Uranium atom⁸. With the present results on $CeCu_{2.1}Si_2$, we now have three heavy-fermion superconductors which show coexisting magnetic ordering with extremely small ordered moments ($0.001 \sim 0.1\mu_B$). This may then be a common feature of the superconductivity in heavy-fermion systems.

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